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14. ABSTRACT
Nuclear isomers can store tremendous amounts of energy for long times - for example, the 31-year metastable excited state of ^{178m2}Hf stores 2.445 MeV per nucleus, or 1.2 GigaJoules per gram. These special states of certain isotopes therefore may prove useful as nuclear batteries, provided that a means is found by which to control (trigger) their energy release upon demand. The concentration of current research is on the use of externally-produced photons to serve as such a trigger. At this point, much research must be conducted to test this process and measure the important physical parameters, from which an evaluation of the feasibility of applications may be conducted. The x-ray generating and support equipment purchased through this award are permitting improved investigations of triggered gamma emission from nuclear isomers and have created, at Youngstown State University's X-Ray Effects Laboratories (XEL and XEL^2), a unique facility for these studies.

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MOTIVATION

Long-lived excited states, or isomers, existing in many atomic nuclei are nature's nuclear batteries. As such, isomers can store tremendous amounts of energy on an atom-by-atom basis, and materials can be created that contain many such atoms. Concentrated into a solid, one gram of an isomeric material can store up to 1.2 gigaJoules, as much energy as more than 100,000 grams of chemicals like gasoline or high explosives, for decades. One of the most attractive aspects is that the stored energy is often released from isomers entirely in the form of high-energy electromagnetic radiation. Since the emission of gamma rays allows the nucleus to de-excite without changing its internal composition, there is no direct transmutation of the nucleus with production of radioactive by-products by the triggered release. Of course, some isotopes do not have stable ground states and this would, following a triggered event, leave residual radioactivity. The ground-state decays themselves could provide a source of additional energy.

A number of possible applications have been suggested for isomers, with a recent concentration on those that would benefit from a burst of gamma radiation, triggered upon demand and leaving a relatively "clean" residue. Possible concepts include:

- Delivery of therapeutic radiation in bursts upon demand directly to the site of tumors,
- Sterilization of biohazards or destruction of chemicals using bursts of radiation,
- Creation of a laser that produces directional beams of coherent gamma rays; a *gamma-ray laser.*

For some applications, the ionizing quality of gamma rays would be most important, for example in damaging unwanted cells or other materials. The technological challenges are greatest in attempting to create a gamma-ray laser, but the result could be a device capable of manipulating matter at the smallest possible scales and delivering energy over great distances.

The promise of such applications, and others presently unforeseen, has motivated considerable research into why isomers exist, how they can be produced and how a release of the stored energy can be triggered. Of course, there is also a great deal of interest motivated purely on the basis of the insight into nuclear structure that isomers provide. Recent studies have suggested that low-energy x rays may trigger gamma emission and presently refined experiments are underway to test those results and to determine the precise energy(ies) needed for the trigger photons and the efficiency for the trigger process. This award has significantly enhanced the ability to perform these measurements, using currently available isomeric sources, by the acquisition of systems for production of trigger x rays that are well-suited to experiments. So equipped, the Youngstown State University X-Ray Effects Laboratories (XEL and XEL^2) comprise unique facilities for the study of triggering of electromagnetic pulses from isomers.

TECHNICAL BACKGROUND

Isomers

There is a key difference between the energy stored and released from isomers and that occurring in traditional fission and fusion reactions. In the latter, the energy is stored in the binding together of the constituent particles within a nucleus, and released by breaking a large nucleus into smaller ones or fusing two small nuclei into a slightly larger one. Either way there is a certainty of radioactive by-products. In isomers, the extra energy is stored in additional motion of the constituent nucleons within the nucleus. A release of isomer energy in the form of gamma rays simply reflects a reduction in the motion of those nucleons. For this reason, it has been said that isomeric energy is more kinetic than nuclear.*

Generally, energy can be stored within nuclei in the motion of the constituent particles or of the nucleus as a whole. The energies and angular momenta of single-particle states intrinsic to a nucleus are governed by the typical rules of quantum mechanics. One particularly important aspect is that most of the isotopes of interest for isomeric materials contain nuclei whose shapes are deformed from spherical. Usually the nuclear shape is prolate, somewhat like a rugby ball, having a unique body-centered axis. It is then possible to define the projection of the angular momentum vector associated with any energy level upon that axis. The quantum number associated with this projection is called K and assumes the primary role in understanding why some excited nuclear states can store such large amounts of energy for very long times [1].

Quantum mechanics dictates that for a nuclear state of specific angular momentum quantum J, the magnitude of the angular momentum vector is given by $\sqrt{J(J+1)}\hbar$ and the magnitude of the projection is $J\hbar \equiv K\hbar$. If the individual motions of the constituent nucleons gives rise, as occasionally happens, to an intrinsic state of large angular momentum, then that vector is oriented near the deformation axis (so-called *deformation aligned*). For example, a J = 16 intrinsic state of the nuclide ^{178}Hf lies at an excitation energy of 2.445 MeV above the ground state. Therefore, this state is characterized by K = 16, showing considerable alignment of the angular momentum vector with the body axis of the nucleus. Decay of this state requires an energy-releasing transition to some lower-lying level, initiating a cascade that eventually reaches the ground state. It is known that only two intrinsic states (other than the ground state) can be found lower than the J = K = 16 level, one with J = K = 6 and the other with J = K = 8. The ground state is characterized by J = K = 0. Thus, in all instances an electromagnetic decay transition from the 2.445-MeV level to other intrinsic states would require an extremely large multipolarity, suggesting a very low transition probability and a long lifetime [2].

Of course, quantum mechanics offers other possibilities for energy (and angular momentum) within a nucleus. For a nucleus with well-defined symmetry, rotation of the nucleus as a whole is allowed. A collective rotation is characterized by a progression of states whose energy and angular momentum follow the pattern typically seen in diatomic molecules. Each intrinsic state, starting with the ground state, provides some base amount of energy and angular momentum to which one or more quanta of rotational energy and angular momentum may be

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* Phrase suggested by J. D. Corey, Sandia National Laboratories.

added. A 'family' of states associated with the same internal motion of nucleons (corresponding to an intrinsic state) is referred to as a band with the intrinsic state being the *bandhead*. The most important feature is that collective rotation is only allowed by quantum mechanics around an axis that is perpendicular to the symmetry (deformation) axis of the nucleus. Thus, more and more rotation, provided the shape of the nucleus is not too disturbed, increases the energy and total angular momentum associated with nuclear states, but does not change the projection of angular momentum on the symmetry axis. All members of the J = K = 0 ground state band (g.s.b.) in ^{178}Hf have K = 0 up to at least J = 20, reflecting that the angular momentum vectors of such levels are oriented along the rotation axis (so-called *rotation-aligned*).

Figure 1 shows the relevant known energy levels of the isotope ^{178}Hf with bands separated visually. Many more levels are known, of course. Due to the presence of the rotational levels, there are a number of states to which the 2.445-MeV, K = 16 level could decay by modest multipolarity transitions. One example is that to the J = 13 member of the K = 8 band. As an electric octupole (E3) transition, it would cause the 2.445-MeV state to have a modest lifetime. There is, however, the additional problem of the required change in orientation of the angular momentum vector, reflected by the change in K. The 2.445-MeV state is strongly deformation aligned, with K = 16. The state to which it could decay electromagnetically with the greatest probability (the aforementioned J = 13 state) corresponds to only K = 8, due to the intrinsic bandhead with J = K = 8. Thus a transition to the J = 13 state requires not only a reasonable change of $\Delta J = L = 3$ in the magnitude of angular momentum of the nucleus, but an even greater change in the orientation. The total $\Delta K = 8$ means that there is an excess orientation change of $\nu = \Delta K - L = 5$. Typically, for nuclei in the $A \sim 180$ region, electromagnetic transition rates are found to be reduced (inhibited) by such orientation changes according to a factor of about f^{ν} with $f \sim 10 - 100$. Thus, the 2.445-MeV state in ^{178}Hf cannot decay even as rapidly as might be expected simply for an E3 transition, leading to a halflife of 31 years. This is an excellent example of a *K isomer*, storing an energy equivalent to 1.2 GJ/gram for a long time. Many other isomers exist, but much research has focused recently on this isomer, referred to as[†] ^{178}Hf^{m2}.

Triggering

The ability to trigger the emission of gamma rays from an isomer like ^{178}Hf^{m2} is the key to utilizing these high-energy density materials for most applications. Scientists and engineers have long sought to control the energy stored with the nucleus but only since the invention of the laser in 1961 have there been any real concepts for triggering bursts of gamma rays without production of radioactive by-products. For much of that time the concentration was specifically aimed at finding a way to produce a gamma-ray laser [3], a task that has not yet been accomplished due to the conflicting demands of intersecting branches of physics. Nevertheless, a large number of potential trigger mechanisms have been proposed and investigated both theoretically and, where possible, experimentally. The primary mechanism of interest has been

[†] The 'm2' indicates that this state is the second metastable level above the ground state, a somewhat arbitrary designation. The intrinsic state having J = K = 8 shown in Fig. 1 has a halflife of 4 s and is thus referred to as the 'm1,' or first metastable. The other excited intrinsic state mentioned in the text is not shown in the figure. It lies slightly higher in energy than the m1 level, has J = K = 6 and a halflife of 78 ns. Although this duration makes the halflife of the state measurable, it is not typically designated by the 'm' superscript.

called triggered (or induced) gamma emission and requires the irradiation of isomeric materials with x rays to initiate the release of energy from the isomers (see the review of Ref. [4]). Of course, for practical purposes it seems that the most attractive trigger events would require the smallest possible energy for the trigger in comparison with the amount of energy contained in the particular isomer.

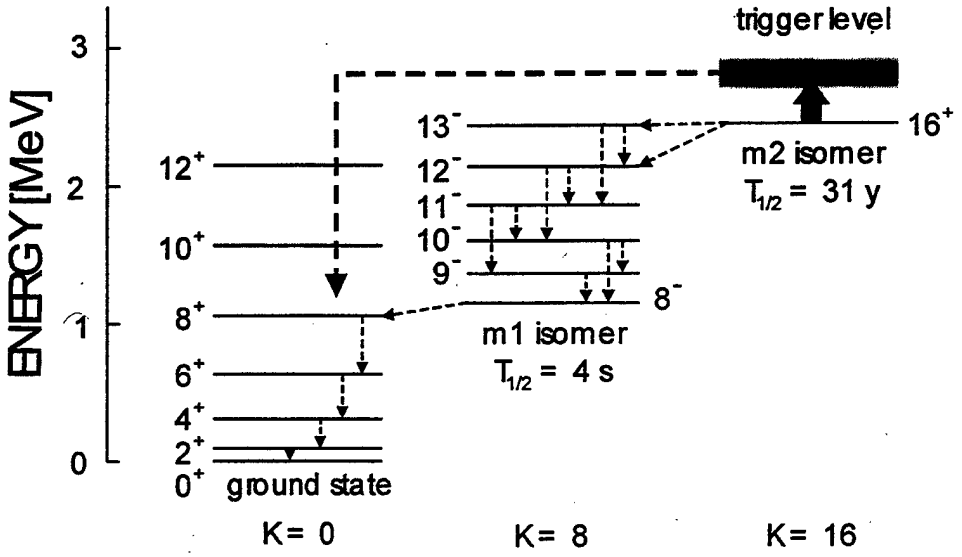

Figure 1: Schematic energy-level diagram that shows relevant known states of $^{178}Hf^{m2}$. The light dashed arrows show transitions that are part of the spontaneous decay cascade of the isomer, which is 'broken' in time as it reaches the 4-s isomer. The heavy upward arrow indicates absorption of an incident photon to an intermediate state that begins the trigger process. The trigger level may then decay by unknown cascade(s) as suggested by the heavy downward dashed arrow.

The trigger mechanism is depicted in schematic fashion in Fig. 1 as a two-step process. An externally-produced x-ray photon incident upon the isomeric material is absorbed by a nucleus that is already in a long-lived metastable state. This absorption serves to excite the nucleus further into a level that must serve to mediate the trigger process, and is therefore referred to as an *intermediate state* or *trigger level*. For the absorption cross section to be large, leading to an efficient trigger reaction, the angular momentum of the intermediate state should be similar to that of the isomeric state. The greatest efficiency would likely result from a dipole transition. Triggering of $^{178}Hf^{m2}$ has been reported in several papers (see the review in Ref. [5]), although there is debate as to whether the mechanism is that of simple photon absorption by the nucleus, or if the nucleus is triggered by a virtual photon due to the NEET process (see the review of Ref. [6]).

In either case, the intermediate state may need to possess a strongly mixed wavefunction, containing components of widely differing K identities, to be excited from the high-K isomer yet decay to low-K members of the ground-state band. This mixing can occur due to near

coincidences of nuclear states that lie in different bands and thus have different K values; nearly coincident states of varied K have recently been experimentally observed [7] in ^{181}Re, but near a 12-μs isomer, too short to be of value for energy storage. Also, if the nuclear interactions for a particular state disturb the axial symmetry of the nuclear shape, such as making it take a triaxial configuration, this would produce a strongly K-mixed wavefunction. Such an occurrence has been detected [8] in ^{186}Os.

Triggered gamma emission using x rays has been demonstrated conclusively for the isomer of ^{180}Ta [9] and suggested for ^{178}Hf^{m2} (see references summarized in Ref. [5]). The isomer ^{180}Ta^{m} is nature's only 'free' metastable state, since its halflife of more than 10^{15} years has insured its survival into the present after its production in stars. The ground state of ^{180}Ta is unstable, having a halflife of 8.2 hours, so all naturally-occurring nuclei of this isotope are in the isomeric state. Unfortunately for applications, that isomer lies only 75 keV above the ground state and it has been shown experimentally [10] that x rays of at least 1 MeV are required to trigger the release of the stored energy. The greatest probability for triggering is for x rays of 2.8 MeV. While unsuitable for applications, this and other studies pointed the way [11, 12] to the possibility of triggering the ^{178}Hf^{m2} isomer, which lies at 2.445 MeV, through an intermediate state expected to lie below 2.8 MeV.

In 1998 a simple experiment was performed [13] in which a dental-style x-ray device was used to irradiate a sample containing 6×10^{14} ^{178}Hf nuclei in the 31-year-lived state. It was reported that the numbers of gamma rays emitted by some transitions in the spontaneous decay cascade of ^{178}Hf^{m2} were increased slightly during the irradiations, while the numbers of gamma rays emitted from a radioactive contaminant, ^{172}Hf, did not change. The low energy of the accelerating potential, 90 kV, implied that triggering gamma emission required x rays of less energy and that the process was quite efficient due to the ability to observe even a small enhancement of the gamma-ray lines with such a weak x-ray source. This and follow-up measurements are the only examples of triggering experiments performed at such low photon energies; in general, the extensive literature of photonuclear reaction studies is confined to higher energies, greater than about 1 MeV and to excitation rather than triggering of isomers [14]. To some degree this has been due to some lack of interest by the mainstream nuclear physics community in low-energy photon reactions (< 2 MeV) and to the lack of suitable x-ray devices to explore this energy range.

Since 1999, there have appeared additional positive reports of triggered gamma emission from ^{178}Hf^{m2}, such as in Refs. [15-19], and several reports of null measurements in Refs. [20-23]. Clearly the question remains open whether low-energy triggering (< 100 keV) can be accomplished for ^{178}Hf^{m2}.

ONGOING RESEARCH PLAN

The critical scientific need regarding triggering of gamma emission from ^{178}Hf^{m2} is to validate low-energy triggering and, if it does occur, to measure the relevant physical parameters. One of the main experimental problems has been the relatively poor statistical accuracy of the reported positive results and so it is necessary for further experiments to employ improved detection techniques. The general techniques available for the observation of triggering from

any isomer are discussed in Ref. [4] and the design and use of a new multi-detector system for triggering studies was reported in Refs. [22, 24]. Here, the emphasis is on the role of the equipment supplied by this DURIP award, which included purchase and installation of additional x-ray generators.

Tests of low-energy triggering (< 100 keV) on $^{178}Hf^{m2}$ were initially motivated by what might be viewed as educated guesses, based on systematics of photonuclear reactions on neighboring isomeric nuclei. Now, with some positive reports of triggering in this range, further studies continue as attempts to validate and improve those measurements. X-ray generating equipment prior to this DURIP consisted of one reconditioned 150-kV radiographic x-ray tube with rotating anode, capable of high-current (up to 100 mA) pulses of maximum 100-ms duration. Longer pulses were possible with lower currents and/or accelerating voltages. This device, switched externally to provide repeated pulse operation, was employed in the initial studies at Youngstown State University [22] using the miniball multi-detector array [24]. However, this tube was not cooled and thus total data acquisition rates were reduced by the need to operate at an operational rate of no more than 0.2 Hz to avoid excessive heating of the rotating anode. It was also necessary to stop irradiations about every 2 hours to allow the tube housing to cool.

Under this DURIP award, it was possible to purchase and install an upgraded 150-kV radiographic x-ray tube with cooler. This tube utilizes the same high-voltage generator as the previous non-cooled tube, minimizing cost. The original non-cooled tube is still available for experiments that might require pulsed operation at higher currents, but the cooled tube will be used more in ongoing experiments of low-energy triggering of $^{178}Hf^{m2}$. The 150-kV accelerating voltage is well-matched to studies in this energy range since this provides significant photon fluxes in the 10- to 20-keV range, while not producing excessive ambient scattering.

In addition to low-energy triggering, so-called mid-energy triggering transitions are accessible to experiments. In an energy range of 100 – 2,000 keV, it is possible to identify specific target trigger transitions from the nuclear spectroscopic level data. The work of Ref. [25] reported decay branches from the 14^- isomer ($T_{1/2}$ = 68 μs) to the 31-year isomer and to levels in the 8^- (4-s) isomer band in ^{178}Hf. Detailed balance indicates that the transition between the 31-year and 68-μs isomers can occur in both directions – thus the 126-keV transition from the longer-lived state to the higher-lying 14^- level will support triggering. This has not, however, been tested experimentally. Also, the recent work of Ref. [26] suggests possible (not definite, as for 126 keV) trigger transitions at 331, 990 and 1,676 keV [5].

Under this DURIP award, a Pantak/Seifert cooled 450-kV x-ray tube was purchased and installed in the X-ray Effects Laboratories at Youngstown State University. This device was chosen for its well-matched capabilities to the needs of mid-energy triggering at 126 and 331 keV. Experiments are underway to perform these tests. Studies of the 990- and 1,676-keV potential trigger transitions require a high-energy x-ray generator, such as a small linear accelerator (linac).

EQUIPMENT PURCHASES

The original proposal submitted to the DURIP program was prepared with two options:

1.	Option 1 - Tunable 450-kV x-ray system consisting of	
	a. Pantak HF450/MXR450/1.5-3.5 X-Ray Unit	$116,519
	b. Pantak High Speed Shutter	$ 6,037
	c. Modular shield enclosure 1	$120,000
	TOTAL	$242,556
2.	Option 2 - Tunable 450-keV, 1-2 MeV X-Ray System, consisting of	
	a. Pantak HF450/MXR450/1.5-3.5 X-Ray Unit	$116,519
	b. Pantak High Speed Shutter	$ 6,037
	c. Reconditioned Varian Linatron 200A X-Ray Linear Accelerator	$150,000
	d. Modular shield enclosure 2	$198,000
	TOTAL	$470,556

Thus Option 2 contained Option 1, plus an additional $228,000. This amount would permit acquisition of the linac needed to investigate the upper end of mid-energy triggering as discussed above, and of a more robust shield enclosure. Award F49620-01-1-0262 corresponded to Option 1. During the period of performance of this award, a second DURIP was awarded (F49620-02-1-0235) for $228,000 – together with the first award, this essentially provided for Option 2. Purchases of equipment under Option 1 was already underway, but it was possible to modify some items to take full advantage of the two awards. Most notably, a quotation was obtained for an upgrade of "Modular shield enclosure 1" to meet the needs of "Modular shield enclosure 2" to provide complete protection from 450-keV x rays. The total amount for the shield enclosure was $140,000, considerably under the expected $198,000 (different company). This made funds available for the purchase of the upgraded 150-kV cooled tube.

The final disposition of purchases under this DURIP award:

1.	Varian G1592/B80 150-kV cooled x-ray tube with HE320 heat exchanger, including installation and calibration	$ 34,285
2.	Pantak/Seifert Isovolt 450 HS 450-kV cooled x-ray tube with standard alarm package and high-speed shutter, including installation and calibration NOTE: Unexpected freight paid from second DURIP award)	$112,950
3.	Modular shield enclosure suitable for 150-keV bremsstrahlung NOTE: Prior to delivery, this was upgraded for 450-keV protection as funded by the second DURIP award.	$ 78,931

4.	Varian Linatron 200A linear accelerator – prepayment to hold machine NOTE: A reconditioned Linatron became available for $150,000 (without chiller) and it became necessary to make a prepayment as a deposit to hold the machine. The remaining cost (with chiller) of $147,510 was funded by the second DURIP award.	$ 16,390
	TOTAL EXPENDITURES	$242,556

EDUCATIONAL IMPACT

Although beyond the typical level of research conducted at comprehensive undergraduate institutions like Youngstown State University, this project has benefited tremendously from the involvement of a number of students at the sophomore, junior and senior levels, and from several different departments. Students Mary Kate Boyle, Yuki Kaneko, Joel Lepak and Rick Toman contributed to every aspect of the planning, design and construction of detector systems and benefited greatly from the unique experience on this project. Ms. Boyle completed a M.S. degree in physics and is now gainfully employed in the private sector on missile defense, while Ms. Kaneko is completing a Ph.D. in astronomy. Mr. Lepak has just entered graduate school in mathematics, seeking a Ph.D. Mr. Toman is gainfully employed in the private sector, involved in the manufacture of laser crystals for inertial confinement fusion devices.

The impact of this project on the education of young scientists has already been considerable and will only expand in the future. Currently five students are involved in the research.

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